Non-spherical aerosol retrieval method employing light scattering by spheroids

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[1] Numerous studies indicate the need to account for particle non-sphericity in modeling the optical properties of dustlike aerosols. The methods for simulating the scattering of light by various non-spherical shapes have rapidly evolved over the last two decades. However, the majority of aerosol remote-sensing retrievals still rely on the Mie theory because retrievals accounting for particle non-sphericity are not as well defined methodologically and are demanding computationally. We propose a method for the retrieval of the optical properties of non-spherical aerosol based on the model of a shape mixture of randomly oriented polydisperse spheroids. This method is applied to angular and spectral radiation measurements from the Aerosol Robotic Network (AERONET) in locations influenced by desert dust. Comparisons with Mie-based retrievals show a significant improvement in dust-particle phase functions, size distributions, and refractive indices. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0933 Exploration Geophysics: Remote sensing; 0994 Exploration Geophysics: Instruments and techniques

1. Introduction

[2] Recently, tropospheric aerosols have been recognized as an important, although highly uncertain, atmospheric constituent influencing the global climate [e.g., *Hansen et al.*, 2000]. The uncertainty in the knowledge of aerosols is explained by difficulties in monitoring spatially- and temporally-variable aerosol properties. Recent progress in both satellite and ground-based remote sensing [e.g. *King et al.*, 1999] enables the scientific community to obtain global aerosol data. However, the accuracy of remote-sensing aerosol characterization is limited by a number of unresolved issues, including the difficulty to model the optical properties of non-spherical dustlike aerosols.

[3] There is sufficient experimental evidence [*Heintzenberg*, 1998; *Volten et al.*, 2001, etc.] that the non-sphericity of desert dust particles can cause scattering properties significantly different from those predicted by the standard Mie theory [*Mishchenko et al.*, 2000]. Although non-spherical scattering has been discussed in the literature for several decades [e.g. *van de Hulst*, 1957], only recent research, aided by progress in computer technology, resulted in practically useful theoretical models. This progress has stimulated efforts in improving the accuracy of aerosol retrievals in the presence of non-spherical particles [e.g., *Kahn et al.*, 1997; *Krotkov et al.*, 1999; *Liu et al.*, 1999]. However, dealing with non-sphericity is not a completely resolved issue, and most aerosol retrieval algorithms are still based on the Mie theory [e.g., *Nakajima et al.*, 1996; *Kaufman et al.*, 1997; *Tanré et al.*, 1997; *Torres et al.*, 1998; *Dubovik and King*, 2000]. Indeed, incorporating non-spherical scattering in remote sensing retrievals is problematic methodologically and technically. Microphoto-graphs of natural aerosols show a great variety of shapes, thus making difficult a realistic choice of a particle shape (or shape mixture) model. Furthermore, all available theoretical methods have certain limitations in their applicability. For example, even the use of the simplest non-spherical particles, spheroids, requires a combination of two methods in order to cover the entire range of aerosol sizes (see below). Also, some remote-sensing observations of dustlike aerosols [*Kaufman et al.*, 1994; *Tanré et al.*, 2001] seem to indicate a significantly lower importance of non-spherical scattering than predicted by laboratory measurements and theoretical models. It is thus clear that the issue of non-sphericity of dustlike particles in aerosol remote sensing needs further analyses.

[4] The aim of this paper is to test a new parameterization of the particle shape in phase function, size distribution, and refractive index retrievals from AERONET [*Holben et al.*, 1998] desert dust observations. This parameterization does not rely on Mie scattering but rather is based on the model of polydisperse, randomly oriented spheroids.

2. Rationale and Solution Approach

[5] AERONET retrievals of aerosol optical properties employ the inversion code that simultaneously fits radiance measurements over a wide angular and spectral range by computations based on an atmospheric radiation model assuming spherical aerosols [*Dubovik and King*, 2000]. As demonstrated by sensitivity studies [*Dubovik et al.*, 2000], this procedure allows the retrieval of both particle size distribution and complex refractive index, as well as such a climatically important aerosol characteristic as single scattering albedo. It also appears that these retrievals are rather sensitive to particle non-sphericity. Specifically, numerical tests have shown that non-sphericity may cause two kinds of retrieval artifacts: (i) a high concentration of very small particles with radii less than 0.1 μm, and (ii) an unrealistically strong decrease of the real part of the refractive index with decreasing wavelength, λ. Both artifacts have been clearly observed in analyses of AERONET data from several locations bordering the Sahara desert [*Dubovik et al.*, 2002]. The agreement between the sensitivity analyses and the actual retrievals motivated us to attempt the development of a retrieval code based on a non-spherical aerosol model.

[6] The sensitivity tests in *Dubovik et al.* [2000] were based on T-matrix computations of scattering by randomly oriented spheroids [*Mishchenko et al.*, 1997]. We decided to implement the same approach in the inversion code of *Dubovik and King* [2000]; this implies forward modeling of the phase function and extinction based on T-matrix instead of previously used Mie simulations. Accordingly, we defined the aerosol single-

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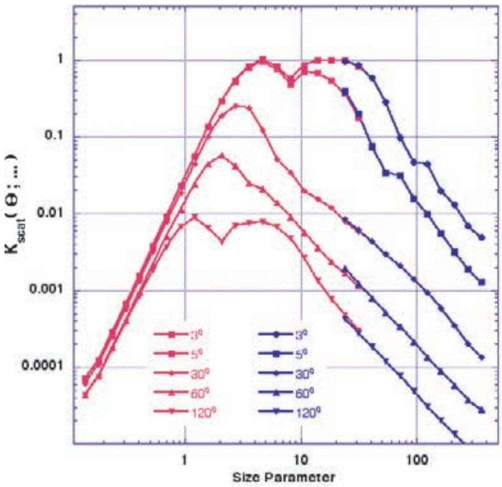

Figure 1. The elements of the kernel matrix $\mathbf{K}_{scat}(\ldots;n, k, \varepsilon_p)$ at different scattering angles for prolate randomly oriented spheroids (ε = 2, n = 1.53, k = 0.003). The red and blue curves show the results obtained with the T-matrix code [*Mishchenko et al.* 1995] and the *Yang and Liou* [1996] method, respectively.

scattering properties as functions of the volume size distribution of randomly oriented, polydisperse spheroids:

$$\tau_{scat}(\lambda)P(\theta;\lambda) \approx \left[\sum_{\varepsilon_p} \mathbf{K}_{scat}(\theta;\lambda;n;k;\varepsilon_p)w(\varepsilon_p)\right]\mathbf{v}, \quad (1)$$

$$\tau_{ext}(\lambda) \approx \left[\sum_{\varepsilon_p} \mathbf{K}_{ext}(\lambda;n;k;\varepsilon_p)w(\varepsilon_p)\right]\mathbf{v}, \quad (2)$$

where τ_{ext} and τ_{scat} denote extinction and scattering optical thickness; $P(\ldots)$ denotes phase function. These linear systems approximate the integral equations for angular light scattering and extinction optical thickness as follows:

$$\int_{r_{min}}^{r_{max}} \left[\int_{\varepsilon_{min}}^{\varepsilon_{max}} \frac{C\ldots(\ldots;r;\varepsilon)}{V(r)} w(\varepsilon)d\varepsilon\right] \frac{dV(r)}{d\ln r} d\ln r \approx \\ \approx \left[\sum_{\varepsilon_p} \mathbf{K}\ldots(\ldots;\varepsilon_p)w(\varepsilon_p)\right]\mathbf{v}, \quad (3)$$

where $\mathbf{v}$ is the vector of the size distribution values $dV(r_i)/d\ln r$ at N_r discrete points r_i. *Dubovik and King* [2000] used N_r logarithmically equidistant size bins $\ln r_i$ ($\ln r_i = \ln r_i + (i - 1)\Delta\ln r$). The elements of the matrices $\mathbf{K}_{ext}(\ldots)$ and $\mathbf{K}_{scat}(\ldots)$ are obtained by integrating the cross sections $C_{ext}(\ldots)$ and $C_{scat}(\ldots)$ of spheroids for each size r_i. Under the simplest assumption of $dV(r)/d\ln r$ being constant within each size bin ($\ln r_i \pm \Delta\ln r/2$), the matrices are computed as

$$\{\mathbf{K}\ldots(\ldots;\varepsilon_p)\}_{ji} = \int_{\ln(r_i)-\Delta\ln r/2}^{\ln(r_i)+\Delta\ln r/2} \frac{C\ldots(\ldots;r;\varepsilon_p)}{V(r)} d\ln r, \quad (4)$$

where r denotes the radius of a volume-equivalent sphere. We adopted the assumption of *Mishchenko et al.* [1997] that the spheroids of all sizes have the same distribution of aspect ratios ε_p, which is represented by the weighting function:

$$\sum_{\varepsilon_p} w(\varepsilon_p) = 1. \quad (5)$$

[7] Equations (1)–(2) are efficient for multiple simulations of scattering by polydisperse aerosols with variable size distributions, because the integration over a particle size range is replaced by using the matrices $\mathbf{K}_{ext}(\ldots)$ and $\mathbf{K}_{scat}(\ldots)$ pre-computed once with high accuracy. This approximation is helpful in improving the time performance of Mie-based retrievals, even though Mie computations are very fast. For the retrieval incorporating non-spherical particles, the use of the above approximations is critical, because direct simulations of non-spherical scattering in the inversion code would be unacceptably time-consuming. The dependence of the matrices $\mathbf{K}_{ext}(\ldots;n, k, \varepsilon_p)$ and $\mathbf{K}_{scat}(\ldots;n, k, \varepsilon_p)$ on the real, n, and imaginary, k, parts of the refractive index can be parameterized by a look-up table covering the entire range of expected values. In the algorithm by *Dubovik and King* [2000], the matrices $\mathbf{K}_{ext}(\ldots)$ and $\mathbf{K}_{scat}(\ldots)$ for spheres were computed at ($N_n \times N_k \times N_r$) grid points. The N_n = 15 equidistant points for n, the N_k = 15 logarithmically equidistant points for k, and the N_r = 22 logarithmically equidistant bins for r sampled the following ranges of the complex refractive index and particle size:

$$1.33 \le n \le 1.6;\ 0.0005 \le k \le 0.5;\ 0.05 \le r \le 15(\mu\text{m}). \quad (6)$$

For values of n and k between the sampling points, $\mathbf{K}_{ext}(\ldots)$ and $\mathbf{K}_{scat}(\ldots)$ were linearly interpolated on the logarithmic scale. This approach allowed fast modeling of atmospheric radiances with accuracy $\sim$1% at the four AERONET sky-channel wavelengths (0.44, 0.67, 0.87, and 1.02 μm).

[8] Thus, we implemented the non-spherical aerosol model via equations (1)–(2) using the matrices $\mathbf{K}_{ext}(\ldots;n, k, \varepsilon_p)$ and $\mathbf{K}_{scat}(\ldots;n, k, \varepsilon_p)$ of integrated cross sections $C_{ext}(\ldots)$ and $C_{scat}(\ldots)$ of randomly oriented spheroids. The integration should be performed for different aspect ratios ε_p and for the $N_r \times N_n \times N_k$ sample points. However, integrating spheroid cross sections over the above ranges of sizes and refractive indices is challenging in practice. For example, even the most advanced T-matrix code [*Mishchenko and Travis*, 1994] becomes numerically unstable for elongated and flattened spheroids ($\varepsilon \sim 2$–2.4) with size parameters x (= $2\pi r/\lambda$) exceeding 40–60. This is a serious limitation compared to the range of x (from $\sim$ 0.3 to 215) suggested by equation (6) for λ from 0.44 to 1.02 μm. Nevertheless, the fact that the rigorous T-matrix approach allows the computations of electromagnetic scattering by spheroids with x up to $\sim$40–60 is very important because for larger x light scattering can be modeled by approximate methods. In this study, we employed the geometric-optics-integral-equation method [*Yang and Liou*,

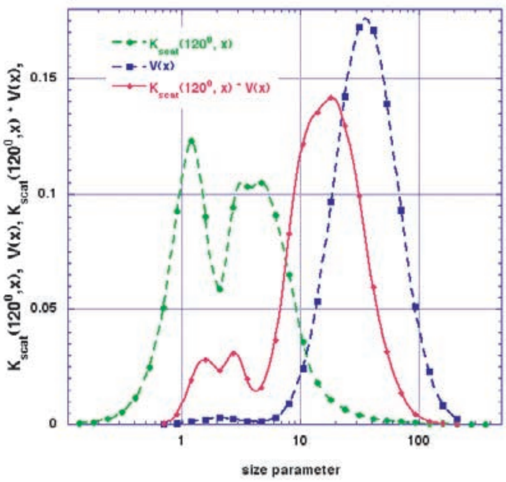

Figure 2. Relative contributions (red solid curve) of different spheroid sizes to light scattering at 0.44 μm (shortest AERONET sky-channel λ) at $\Theta \sim 120^0$ (the scattering angle range of maximal differences between spheres and spheroids). Analogous tendencies were observed for other Θ and for τ.

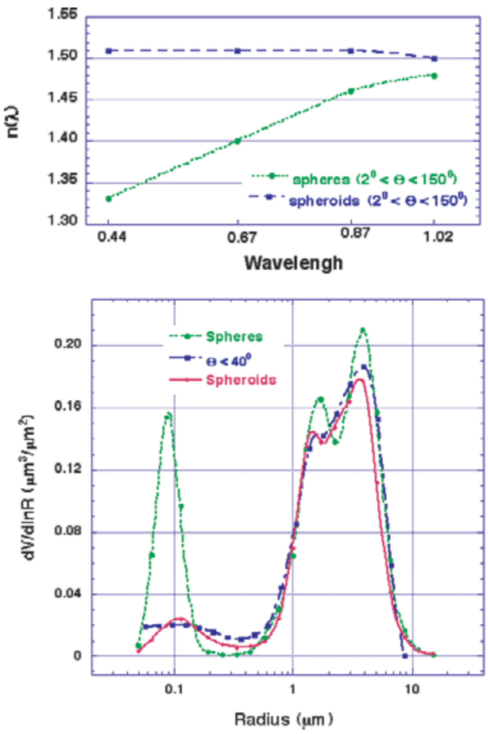

Figure 3. The comparison of desert dust properties retrieved using spheroid and Mie scattering models. (Persian Gulf, May 1999).

1996], in which the ray-tracing procedure is used to find the field on the particle surface. The latter is then transformed to the far field using the equivalence theorem. This approach is applicable to significantly smaller particles than the conventional geometric optics method and provides accurate $P(\Theta)$ and τ for spheroids with x larger than $\sim$30–40. Thus, supplementing the T-matrix method by the geometric-optics-integral-equation technique, we can generate the kernel matrices for spheroids in a wide range of x. Figure 1 illustrates the elements of $\mathbf{K}_{\mathrm{scat}}(\ldots;n,\ k,\ \varepsilon_p)$ calculated using these two codes and shows good agreement for the range of size parameters where both codes are applicable ($x \sim 30$–50).

[9] The very long computational time required for simulating the kernel matrices at all the $N_r \times N_n \times N_k$ sampling points was a serious problem. The method of *Yang and Liou* [1996] integrates numerically the scattering by each single particle over at least 20,000 orientations. This integration was performed at least 10 times for each size bin ($\ln r_i \pm \Delta \ln r/2$) in the equation (4) integration over sizes. The T-matrix method is based on the analytical averaging over spheroid orientations and, therefore, is very fast. However, for x and/or ε close to the numerical instability thresholds, the T-matrix computations also slow down sharply.

[10] According to our estimates, producing kernel matrices for the ranges of equation (6) ($x \sim 0.3$–215) would take about a year or even more, and most of the computational time would be spent for particles with $x > \sim 10$. Therefore, before starting such a long computation we needed to verify the necessity to account for very large particles. For example, *Mishchenko et al.* [1997] limited their dust modeling simulations by $x \sim 50$. We analyzed the contributions of desert dust particles of different sizes to the radiances measured by the AERONET. The results depicted in Figure 2 show that in order to account for $\sim$99% of the actual scattering signal it is necessary to consider aerosol particles with sizes up to at least $x \sim 100$–120. In the analysis we used the desert dust distribution [$\mathbf{V}(\mathbf{x})$ in Figure 2] with the largest median radius of the coarse mode $r_v(\mathrm{coarse}) \sim 2.54$ μm, as observed by AERONET in the Persian Gulf [*Dubovik et al.*, 2002]. Some observations [e.g., *Li-Jones and Prospero*, 1998; *Arimoto et al.*, 1997] indicate size distributions with median radii (for $dV/d\ln r$) up to 5 μm and even larger. Therefore it appears critical to assume the potential presence of particles larger than $x \sim 100$–120 as suggested by Figure 2, and so we relied on the size range of equation (6) covering r up to 15 μm. This required only two points with $x_i > 100$ (for $\lambda = 0.44$ μm) with the chosen logarithmically equidistant bins [$\ln r_i = \ln r_i + (i - 1)\ \Delta \ln r$].

[11] Hence, in spite of the long computing time, we have calculated $\mathbf{K}_{\mathrm{ext}}(\ldots;n,\ k,\ \varepsilon_p)$ and $\mathbf{K}_{\mathrm{scat}}(\ldots;n,\ k,\ \varepsilon_p)$ for randomly oriented prolate and oblate spheroids with ε_p from 1.2 to 2.4 (with step 0.2) and for the n, k, and r values given by equation (6).

3. Results and Discussion

[12] The application of the modified retrieval method based on the spheroidal particle model to AERONET measurements showed clear improvements in the retrieved optical properties of desert dust compared to those derived using the assumption of spherical particles. As Figure 3 illustrates, using spheroids removes both the artificially increased fine particle mode and the unrealistic spectral dependence in the real part of the refractive index. It is important that the retrieved $dV/d\ln r$ of spheroids agrees well with $dV/d\ln r$ retrieved from sky-radiance aureole ($\Theta < 40°$), were the effects of non-sphericity are minimal. For a more convincing illustration, we supplemented the aureole retrieval by the independent method of *Nakajima et al.* [1996], which uses Mie scattering with presumed n and k (we used the values obtained from the spheroid retrieval). Figure 4 shows that $P(\Theta)$ retrieved using spheroids exhibits significant differences with both retrievals that used Mie scattering.

[13] The single scattering albedo and $k(\lambda)$ (not displayed) retrieved using sphere (for $\Theta < 150°$) and spheroid models agreed within retrieval accuracy, as expected by *Dubovik et al.* [2000] (not shown). It is interesting to note that for the fine-mode-dominated aerosols (such as biomass burning and urban/industrial particles) both inversions (based on spheroid and on sphere scattering) resulted in essentially the same properties. For the retrievals shown at Figures 3 and 4 we assumed a shape distribution similar to the model of *Mishchenko et al.* [1997] but with a slightly narrower range of ε_p. Specifically, we used an equiprobable distribution of prolate and oblate spheroids with aspect ratios ε_p ranging from 1.6 to 2.2. This shape distribution was chosen in a series of test retrievals. We inverted many real AERONET observations of desert dust assuming spheroids of single shape [i.e. no ε_p averaging in equations (1)–(2)]. We found that the retrieval artifacts attributed to ignoring particle non-sphericity were significantly reduced

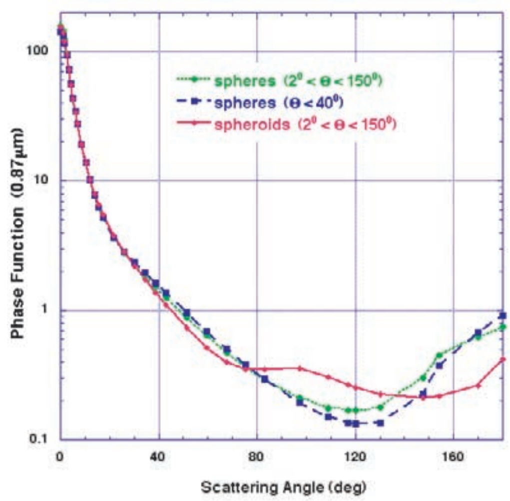

Figure 4. Comparison of desert dust phase functions retrieved using the spheroid and Mie-based scattering models.

if we used single-shape spheroids with ε_p in the range from 1.8 to 2.0, whereas for ε_p both smaller than 1.6 and larger than 2.2 the artifacts were strongly pronounced. Also we found that using equally mixed prolate and oblate spheroids with ε_p from 1.6 to 2.2 resulted in superior retrievals compared to those obtained using spheroids of any single shape or other tested mixtures of shapes.

[14] In order to verify how representative the retrieval improvements in Figures 3 and 4 were, we processed a large volume (> 3000 cases) of AERONET data collected at several desert dust sites. Significant improvements (similar to those demonstrated in Figures 3 and 4) were obtained in ~90% of all cases. In ~10% of cases, the retrieval artifacts mentioned above remained pronounced, which can be potentially explained by several reasons. For example, dust particles are not ideal spheroids and dust may have scattering properties not exactly reproducible by the spheroid model. However, in order to definitively conclude that we are facing such limitations, all aspects of the spheroid model should be explored. For example, the aspect-ratio distribution was fixed in our method, while in reality it may change. Also, in order to afford the computation of the kernel functions, we used moderate accuracy criteria in the integration in equations (1)–(2), resulting in phase function modeling errors of up to 5%. For example, the retrieval artifacts appeared more frequently when sky-radiances were measured only up to scattering angles 60–80°, i.e. in the Θ-range where non-spherical scattering effects are moderate. In such cases our modeling accuracy can be unacceptable. Also, our forward-scattering model was based on a scalar radiative transfer code, whereas it is conceivable that for non-spherical particles even the computation of intensity requires the full accounting of polarization effects.

[15] However, the significant improvement in the 90% of dust retrievals clearly indicates the importance of taking into account the effects of nonsphericity in the retrieval algorithm. Indeed, using the spheroid model we were able to derive more accurately particle size, complex refractive index, and phase function from atmospheric radiances measured in a wide scattering-angle range (from 3° to ~150°). The improvement in the derived phase functions is probably the most important, because a correct $P(\Theta)$ cannot be simulated even from the correct size distribution (e.g. obtained from measurements at $\Theta < 40°$) and known refractive index without addressing the issue of particle shape.

4. Conclusions

[16] We have employed the model of dustlike aerosol particles as polydisperse, randomly oriented spheroids for retrieving aerosol optical properties from remote measurements of atmospheric radiances. The application to AERONET measurements taken at several dust-dominated locations has shown significant improvements in the retrieved size distribution, refractive index, and phase function. Nevertheless, this study is only our first attempt to explore the implications of non-spherical particle scattering for desert dust characterization, and several aspects of the retrieval approach are planned to be improved. For example, we plan to address the possibility of optimizing particle aspect- ratio distributions for each retrieval and to improve the accuracy of computing the spheroid kernel matrices. The validation of the retrievals by independent field measurements will also be an important factor in further justification and improvement of the presented non-spherical aerosol retrieval method.

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